Third-order optical autocorrelator for time-domain operation at telecommunication wavelengths

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We report on amorphous organic thin films that exhibit efficient third-harmonic generation at telecommunication wavelengths. At 1550 nm, micrometer-thick samples generate up to 17 μW of green light with input power of 250 mW delivered by an optical parametric oscillator. This high conversion efficiency is achieved without phase matching or cascading of quadratic nonlinear effects. With these films, we demonstrate a low-cost, sensitive third-order autocorrelator that can be used in the time-frequency domain. © *2004 American Institute of Physics*.
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Driven by the development of all-optical networks with increasing bandwidths, ultrafast lasers are gaining in power and portability. Mode-locked erbium doped fiber lasers that emit subpicosecond pulses at telecommunication wavelengths have been demonstrated with average power of several hundred mW.[1] Other widespread applications for the use of such lasers include terahertz imaging,[2] nonlinear sampling,[3] optical coherence tomography,[4] and multiphoton imaging.[5] With the development of these high-power ultrashort laser sources, there is an increasing need for practical, accurate solutions for pulse diagnostics.

For the characterization of femtosecond and picosecond pulses, techniques based on second-order nonlinear processes are widely used in noncollinear or collinear geometries, providing intensity and interferometric autocorrelations, respectively.[6] Other second-order autocorrelation techniques are based on two-photon absorption processes in semiconductors.[7] These processes are usually sensitive enough that low energy pulses can be characterized, but suffer from limited sensitivity to a particular pulse shape. For instance, second-order autocorrelations, being symmetrical functions, do not provide information on the pulse asymmetry. Furthermore, the second-harmonic signal is generated in thick crystals in a geometry that satisfies phase-matching conditions. Hence, the process is polarization sensitive and has a limited wavelength tunability range.

Third-order nonlinear processes have been recognized to be superior to second-order ones but are limited in sensitivity due to the lack of materials with strong third-order nonlinearity and transparency. Most of these third-order techniques are currently implemented with fused silica. Third-order autocorrelation can be implemented by using three-photon absorption in photodiodes.[8,9] But the sensitivity of these techniques is limited when simultaneous measurement of the spectrum of the nonlinear signal is required to retrieve information on the phase of the pulse, like in frequency-resolved optical gating (FROG) techniques.[10] Recently, Third-harmonic generation (THG) was demonstrated in poly(*p*-phenylenevinylene) (PPV),[11] and applied to autocorrelation. However, these films require conversion from a precursor at elevated temperature and are known to be unstable under illumination.

In this letter, we report on amorphous organic thin films that exhibit efficient THG at telecommunication wavelengths when excited with unamplified ultrafast pulses. Microwatt-level THG signals are used to generate third-order autocorrelation traces for characterization of the laser pulses. The third-order autocorrelator we demonstrate has several advantages: the third-harmonic signal is strong enough that it can easily be detected with a standard unamplified silicon detector which makes it practical and low cost; with such high signal levels, simultaneous measurement of the spectrum is

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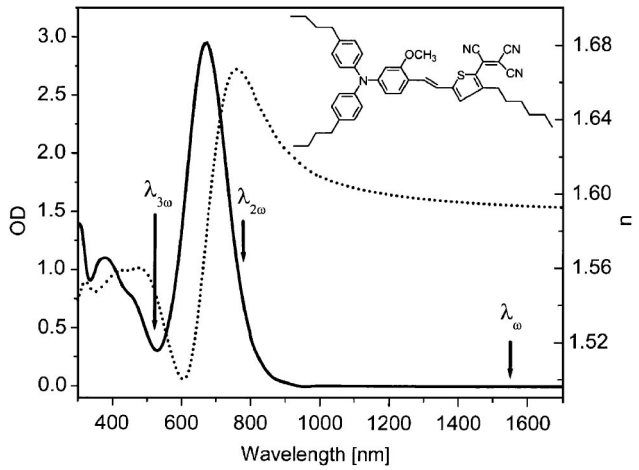

FIG. 1. Absorption spectrum (solid line) measured in a thin sample and calculated index dispersion (dotted line). Inset: Molecular structure of the nonlinear chromophore. Arrows indicate the frequencies for the fundamental, two-photon resonance, and third harmonic.

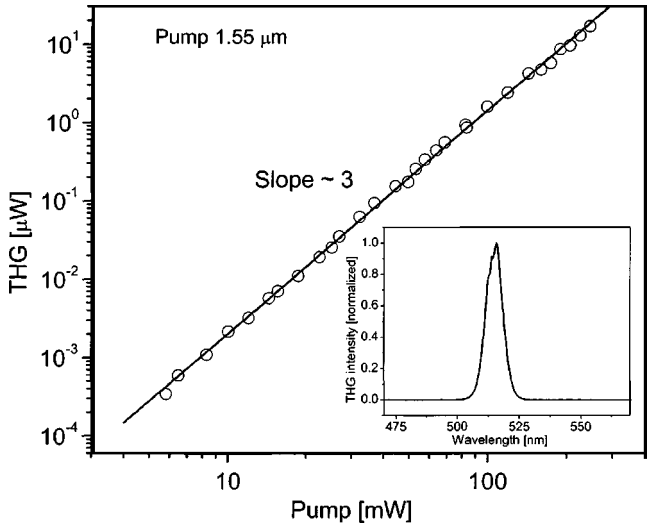

FIG. 2. Third-harmonic signal power as a function of the input fundamental power measured in a 10-μm-thick sample at 1550 nm. Inset: Spectrum of the third-harmonic signal measured behind the sample with a fiber spectrometer.

possible with compact portable low-cost fiber spectrometers; the sample thickness is thin (a few micrometers) which limits potential distortions caused by geometric, dispersive, and phase-mismatch effects; the material is fully isotropic, which makes it completely polarization insensitive; high THG signals are produced without the use of phase matching or cascading of quadratic nonlinear effects, which provides a broad wavelength tuning range.

For our experiments, we used an optical parametric oscillator that emits pulses at a rate of 82 MHz with typical duration of 90 fs that are tunable from 1.4 to 1.6 μm. Autocorrelation measurements were performed with a standard Michelson interferometer setup in which the relative optical pass length between the arms of the interferometer was modulated by a PZT crystal at frequency of 1 Hz. The two copropagating beams were focused onto an organic thin film sandwiched between two glass slides with a 4 cm lens. The third-harmonic autocorrelation signal was collected by a second lens, separated from the fundamental by means of a color filter, and detected by a standard nonamplified silicon detector. For the spectrally resolved experiments we used a portable fiber spectrometer.

The organic thin film is a polymer composite containing the push–pull chromophore [2-tricyanovinyl 3-hexyl-5(4-*NN′*diphenyl-4-dibutyl) vinyaniline thiophene][12] the molecular structure of which is shown in the inset of Fig. 1. The chromophore is doped into a polymer host. For the studies presented here, the matrix was polystyrene and the doping level was 20 wt %. Comparable results can be obtained in other polymer matrices and at different doping levels. The samples were prepared in air by first dissolving the guest chromophore and the host polymer matrix in chloroform and stirring. Then, the solvent was evaporated using a rotor vapor and yielded a solid material. The solid polymer composite was sandwiched between two glass slides and melted on a hot plate at 170 °C. The thickness of the sample was controlled by calibrated spacers. The samples were then encapsulated by applying epoxy around the edges of the glass slides. The films had excellent optical quality, showing negligible light scattering in the visible and near infrared (NIR) and shelf lifetimes of several months.

Figure 1 shows the measured linear absorption spectrum of a thin organic film made from the composite by spin coating. The arrows indicate the spectral position of the fundamental laser signal (1550 nm in this case), the wavelength corresponding to twice the photon energy, and that of the third-harmonic signal, respectively. Under optical excitation with a single beam at wavelengths used for telecommunications (1.4–1.6 μm), a strong visible third-harmonic signal is observed by the naked eye in standard laboratory lighting conditions. The strong third-order harmonic signal is due to relatively low absorption of the sample on the high energy side of the absorption band where the signal is generated. This reduces reabsorption of the nonlinear signal. Figure 1 also shows the dispersion of the refractive index calculated from the absorption spectrum using Kramers–Kronig relations and by matching the refractive index of the film that was measured at 1550 nm with a prism coupler (Metricon, model 2010).

To characterize the nonlinearity of our films we measured the third-harmonic signal power in a single beam experiment as a function of the pump power. Figure 2 shows the results measured in a 10-μm-thick film. It clearly shows that the signal has cubic dependence on the incident power. At the maximum pump power of 250 mW, a third-harmonic signal of 17 μW was generated. An estimate of the magnitude of the third-order susceptibility of our films was made in plane wave approximation, yielding a value of $\chi^{(3)}=0.82\times10^{-19}$ (m/V)^2 (0.62×10^{-11} esu).

Figure 3 shows a real-time third-order interferometric autocorrelation trace using a 12-μm-thick film with an input pump for the autocorrelator of 60 mW, which after Fresnel losses in the beam splitter is reduced to 22 mW at the ouput. In Fig. 3 we clearly see a signal to background ratio of 32:1 distinctive of third-harmonic generation autocorrelations.[13] Assuming that the pulses have a Gaussian shape, the deconvolution of the third-order autocorrelation yields a pulse of 93 fs [full width at half maximum (FWHM)]. The spectral width of the incident pulses was 38 nm (FWHM), indicating that they were nearly Fourier transform limited. For comparison purposes, we also generated a second-order autocorrela-

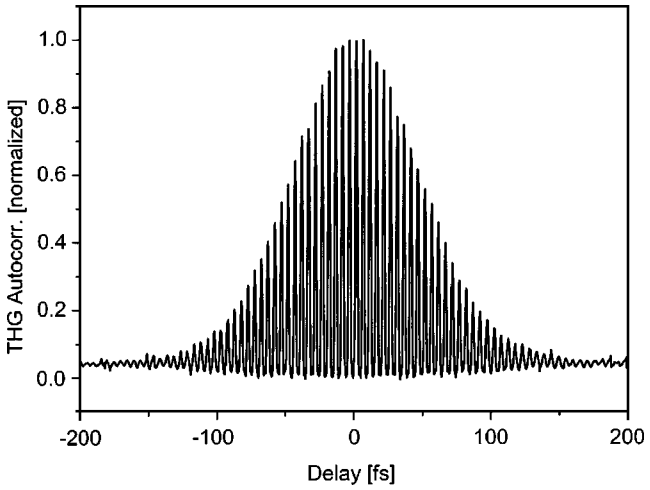

FIG. 3. Interferometric autocorrelation trace measured from the third-harmonic signal.

tion by replacing the organic film with a 500-μm-thick BBO crystal. An identical pulsewidth was obtained.

The advantages of third-order autocorrelation techniques over second-order techniques have been previously discussed[9,10,13] and solutions based on three-photon absorption in photodiodes[9] provide a means by which to characterize asymmetric pulses and remove direction-of-time ambiguity. Unfortunately, they provide only information on the pulse's amplitude not its phase. For complete characterization of short pulses, full knowledge of both is required. This can be done through time-frequency domain measurements such as FROG. Such pulse measurement methods usually require spectrometers coupled to expensive highly sensitive detector arrays. With the third-order autocorrelator that is described here, the nonlinear signal power is sufficient that its spectrum can be measured with a low-cost spectrometer such as a compact portable fiber spectrometer. The inset of Fig. 3 shows an example of such an experiment and demonstrates its feasibility.

In conclusion, nonlinear optical autocorrelation by use of efficient third-harmonic generation in an organic polymer composite was demonstrated. An efficient third-harmonic signal could be produced in μm-thick films by choosing the nonlinear chromophore such that the material is transparent at the photon energy of the incident pulse, strongly absorbing at twice the photon energy to provide a two-photon resonance effect to the nonlinearity, and weakly absorbing at the third harmonic to reduce reabsorption. The autocorrelator we demonstrated combines low cost, portability, high sensitivity, and polarization insensitivity. Furthermore it can be used to characterize pulses from 1.4 to 1.6 μm which makes it useful for the characterization of pulses for telecommunications. More important, its high sensitivity makes it suitable for full characterization of ultrafast pulse using a time-frequency domain technique such as (FROG)[10] using low-cost portable fiber spectrometers. Such experiments are currently underway.

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